



# Aridity and geochemical drivers of soil micronutrient and contaminant availability in European drylands

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## Abstract

Dryland soils provide different societal and environmental services, such as food supply and biodiversity support. In Europe, most of the dryland areas are devoted to agriculture. In the next decades, both European and worldwide drylands are expected to suffer with increased intensity due to the expected climate change-derived rise in aridity. Many studies have focussed on aridity-induced changes in major nutrients in drylands, but little is known of the impact of environmental and biogeochemical factors on micronutrients with critical roles in life, and as inorganic contaminants with ecotoxicological implications. We analysed and explored drivers of total and available concentrations of micronutrients (Co, Cu, Fe, Mo, Mn, Ni and Zn) and contaminants (As, Cd and Pb) in 148 soil samples collected from European drylands covering a wide range of aridity and of other geochemical parameters. The availability of micronutrients increased with their total content, decreased with pH and was enhanced by organic C content. Aridity decreased the availability of Fe, a key element in human diet. Our findings also highlight the scarcity of this micronutrient in European drylands, as well as of some other important micronutrients like Zn and Mo in agricultural soils. Total content was the main driver of the availability of Cd and Pb, and organic matter exerted synergistic effects on contaminant release. Our data show the need for precise management practices to be incentivised by agricultural and environmental policies in order to ensure micronutrient supply and avoid contamination, thus maintaining adequate levels of agricultural productivity and simultaneously preserving dryland ecosystems.

## Highlights

- Drylands are important for food production in Europe and sensitive to climate change.
- The occurrence of metals in European Union dry soils and the drivers influencing them were studied.

- Some micronutrients (Fe, Mo and Zn) were scarce while contaminants were abundant.
- SOC, pH and clays were the main drivers of element availability; aridity reduced Fe.
- Agricultural practises are needed to ensure nutrient supply and prevent contamination.

**KEYWORDS**

agriculture, clay, deficiency, metals, soil organic carbon, soil pH

## 1 | INTRODUCTION

Soils are key to supporting healthy ecosystems and supply goods and services, and are thus pivotal for achieving the UN's Sustainable Development Goals (Keesstra et al., 2016; Lorenz et al., 2019). However, soils are vulnerable to human-induced effects such as climate change, contamination, erosion and land use (e.g., agriculture and industry) (Jie et al., 2002; Lal, 2009). These factors are altering soil parameters, such as water availability and temperature (Colantoni et al., 2015; Gao & Giorgi, 2008), and soil chemical composition, including organic matter content, pH and texture (Delgado-Baquerizo et al., 2013; Jiao et al., 2016). Soil quality, namely the ability of soils to provide functions (e.g., element cycling [Bünemann et al., 2018]) and support services (e.g., food production [Bünemann et al., 2018]) is affected by both soil parameters and human-induced effects (de Paul Obade & Lal, 2016). In particular, intensive exploitation has strongly affected the physico-chemical properties of soils in the European Union (EU) over the last decades (Okpara et al., 2020; Tóth et al., 2008), decreasing soil quality. In addition, this negative effect on soils can be exacerbated by urbanisation and climate change, this ultimately conditioning future agricultural production (Gardi et al., 2015).

Drylands—regions characterised by a water deficit—represent a key ecosystem in the EU and cover an important area of Southern Europe (Právělie, 2016), and are mainly devoted to agriculture (“EUROSTAT—Land use statistics,” Eurostat, 2015). In the ongoing context of environmental changes, EU dryland soils are expected to suffer from increases in aridity in the near future (Právělie et al., 2019). However, the consequences of increasing aridity on soil properties and ecosystem functioning remain underexplored (Delgado-Baquerizo et al., 2013). This is particularly true for key soil functions, such as micronutrient provision to, and contaminant retention from, biota (i.e., plants and animals) (Banwart et al., 2019). Unlike the drivers of the

availability of macronutrients, such as carbon (C), nitrogen (N) and phosphorus (P), the factors governing the availability of metallic micronutrients, such as iron (Fe), zinc (Zn), manganese (Mn), molybdenum (Mo) and copper (Cu), and metallic contaminants, such as lead (Pb), cadmium (Cd) and arsenic (As), have not been extensively studied at a large scale. Micronutrients are essential for life and basic requirements for key biological functions such as photosynthesis, respiration, reproduction, enzyme composition and N fixation (Barron et al., 2009; Broadley et al., 2012), while contaminants represent a risk for life if present and/or assimilated over certain thresholds (Bååth, 1989; Moreno-Jiménez et al., 2011; Nagajyoti et al., 2010). Thus, unravelling the influence of environmental and geochemical parameters on soil micronutrient availability is crucial to manage agrosystems sustainably and preserve natural resources.

The availability of soil micronutrients and contaminants to plants and soil organisms is known to be affected by soil pH, organic matter and clay contents; for instance, high pH and high clay content reduce the transfer of Fe or Zn to plants, but high organic matter enhances it (Alloway, 2009; Hansen et al., 2006). It is also known that aridity may reduce global soil Fe, Zn, Cu and Mn availability (Moreno-Jiménez et al., 2019). In the agricultural context, Fe and Zn are known to be deficient nutrients in human diets worldwide (Ramakrishnan, 2002). Basic soils with low organic matter, which are typical of drylands, are known to exacerbate the supply limitation of Fe and Zn to crops (White & Broadley, 2005; White & Zasoski, 1999; Fageria & Stone, 2008; Ryan et al., 2013). As a result, the Food and Agriculture Organisation considers Fe and Zn deficiency as a main threat for human health and calls for crop biofortification of these micronutrients as a possible palliative counteraction (Thompson & Amoroso, 2010). Likewise, contaminant transfer from soil to crop, and particularly the occurrence of As, Cd and Pb in staple foods, is a main concern in the EU (CONTAM, 2010; EFSA, 2012, 2010), and their soil biogeochemistry is also expected to be influenced by

geochemical and environmental parameters, similarly to micronutrients (Tack, 2010). In a context of climate change models that forecastg an increase in aridity in dryland areas, such as those in Southern Europe (Právělie et al., 2019), the consequences for soil metallic micronutrients and contaminants are highly uncertain. Reducing this uncertainty is critical for agricultural planning and environmental preservation.

Here, we examine the availability of metallic micronutrients (Cu, Fe, Mn, Mo, Ni and Zn) and contaminants (As, Cd and Pb) in a large pool of soil samples from drylands in Europe (Lindsay & Norvell, 1978). We investigate the correlation between nutrients, geochemical drivers (i.e., soil metal concentration, clay content, pH and organic C) and aridity. Finally, we discuss the implications of our results in the context of the EU agriculture and environment political scenario. Despite focusing on the south of Europe, our results could be extended to a rising number of agricultural regions worldwide, not just in drylands, showing similar soil properties (pH neutral to basic and low organic C content) and affected by increased aridity.

## 2 | MATERIALS AND METHODS

### 2.1 | Soil survey

Soil samples were collected from 148 sites located in drylands of six European countries (Portugal, Spain, Italy, Hungary, Greece and Malta; Figure S1) (Právělie, 2016) in 2009 and 2012, following a standardised sampling protocol as part of the European soil survey LUCAS (see Orgiazzi et al., 2018 for details). Drylands are characterised by water deficit, with values of Aridity Index (AI, ratio of precipitation to potential evapotranspiration)  $<0.65 \text{ mm mm}^{-1}$ .

The soils included 10 classes from the World Reference Base (95 Luvisols, 17 Vertisols, 11 Calcisols, 6 Leptosols, 5 Phaeozems, 4 Chernozems, 3 Cambisols, 3 Fluvisols, 2 Arenosols and 2 Kastanozems; IUSS Working Group WRB, 2006) and the types of land uses were 63% agricultural, 20% grasslands, 10% bare lands, 5% forests and 1% shrublands. Samples were grouped into two categories attending to soil management intensity: croplands (including agricultural lands and part of grasslands) and non-croplands (rest of the uses). Not all grassland sites were included in “cropland” category as only a few (9 out of 30) showed clear signs of management and grazing based on LUCAS land cover classification (Eurostat, 2009). The study sites covered a wide gradient of soil geochemical (clay content, organic C and pH) and environmental (aridity) variables typical of European drylands (Figure S2).

### 2.2 | Environmental parameters and soil chemical analysis

The location of soil sampling was registered through global positioning system coordinates and reported in the European Soil Data Centre (Panagos et al., 2012). This allows us to visualise and further analyse them. The AI of the sites was extracted from the CGIAR-CSI v2 database (<http://www.cgiar-csi.org/data/global-aridity-and-pet-database> [Zomer et al., 2008]), which is based on climatic data from WorldClim v2 (Hijmans et al., 2005), and transformed to Aridity (1-AI). Soil pH, organic C and clay content were derived from LUCAS database (details about applied methodology in Orgiazzi et al., 2018).

We analysed two relevant fractions of metals in soils: total and diethylenetriaminepentaacetic acid (DTPA)-extractable (or available) concentrations. Total metal concentration was analysed in aqua regia ( $\text{HCl}/\text{HNO}_3$ ) extracts after heating at  $200^\circ\text{C}$  (MarsExpress) and subsequent filtration through a 42 Whatman filter. The DTPA-extractable element concentration was extracted from soil with 0.005 M DTPA, 0.01 M  $\text{CaCl}_2$  and 0.1 M triethanolamine (pH 7.3), shaking for 2 h at room temperature ( $20^\circ\text{C}$ ) and filtering the supernatant through a 42 Whatman filter (Lindsay & Norvell, 1978). We used the DTPA method as indicator of available pool of metals in soils because this technique is extensively used for these purposes in neutral to basic soils, as is the case of most of the studied soils (de Santiago-Martín et al., 2015; Liang & Karamanos, 1993; Lindsay & Norvell, 1978). Total and DTPA extractions were analysed by inductively coupled plasma mass spectroscopy (Perkin-Elmer NexION 300XX). A certified reference material (CMR048-050) was used to control the quality of soil metal assessment, and the recoveries for the studied elements (As, Cd, Co, Cu, Fe, Mn, Mo, Ni, Pb and Zn) ranged between 86% and 103%.

### 2.3 | Data processing and statistical analyses

We used confirmatory path analysis (CPA) to examine the relative importance of aridity and soil variables on soil micronutrients and contaminants, total and available concentrations. This analysis allows us to assess complex dependencies among several variables, including the evaluation of direct and indirect effects on our response variable of interest (availability) (Shipley, 2009). In these models, we included both soil parameters (pH, clay content, organic matter and total metal concentration), which are well-recognised modulators of metal geochemistry and can have a direct effect on soil metal availability (McBride, 1989; Sauvé et al., 2000); and

aridity as a recently reported driver of soil metal geochemistry (Moreno-Jiménez et al., 2019). All variables were transformed to attain normality and standardised before being included in the models. Furthermore, as variables included in the analysis presented significant spatial clustering ( $p$ -value for Moran I < 0.05 for all variables), in each model we included a spatial autocorrelation random effect based on the geographical distance between all study sites. We tested a total of 12 models, one model for each individual metal (10 models in total), plus two models including composite indices for both total and available soil micronutrients (Co, Cu, Fe, Mn, Mo, Ni and Zn) or contaminants (As, Cd and Pb), built by adding total or available values for each individual element included in the indices. To standardise the contribution of each element to the composite indices, we used molar concentrations (i.e., we divided the mass concentration of each metal by its atomic weight before being added).

We built our CPAs considering an a priori model that included all the potential relationships between the variables (Figure S3). Then, the model was simplified by doing a stepwise variable selection by removing at each step the path with less explanatory power, until the best model based on Akaike and Bayesian Information criteria was found (Shipley, 2009). In addition, we calculated the direct and the indirect effects (i.e., effects mediated through direct effects on other variables affecting the endogenous variable of interest) of environmental drivers on soil metal content and availability. Direct effects are calculated as the sum of the effects of all variables directly affecting the variables of interest, while indirect effects are calculated as the sum of all indirect effects, calculated as the product of the effects for all variables involved in each indirect path (i.e., in the path  $A \rightarrow B \rightarrow C$ , for the variable  $C$  direct effect is  $B \rightarrow C$  while indirect effect is  $(A \rightarrow B) * (B \rightarrow C)$ ). Finally, to identify the predominant drivers for soil metals, we calculated the total effect of environmental variables as the sum of both their direct and indirect effects. All data processing and statistical analyses were conducted with the piecewise *SEM* package (Rosseel, 2012) for R.3.5.1 (Team, 2014).

### 3 | RESULTS

#### 3.1 | Concentration and availability of metals in European dryland soils

The total concentration of all micronutrients was below average values commonly found in soils (He et al., 2005; Lindsay, 1979) (Figure 1). In contrast, the total

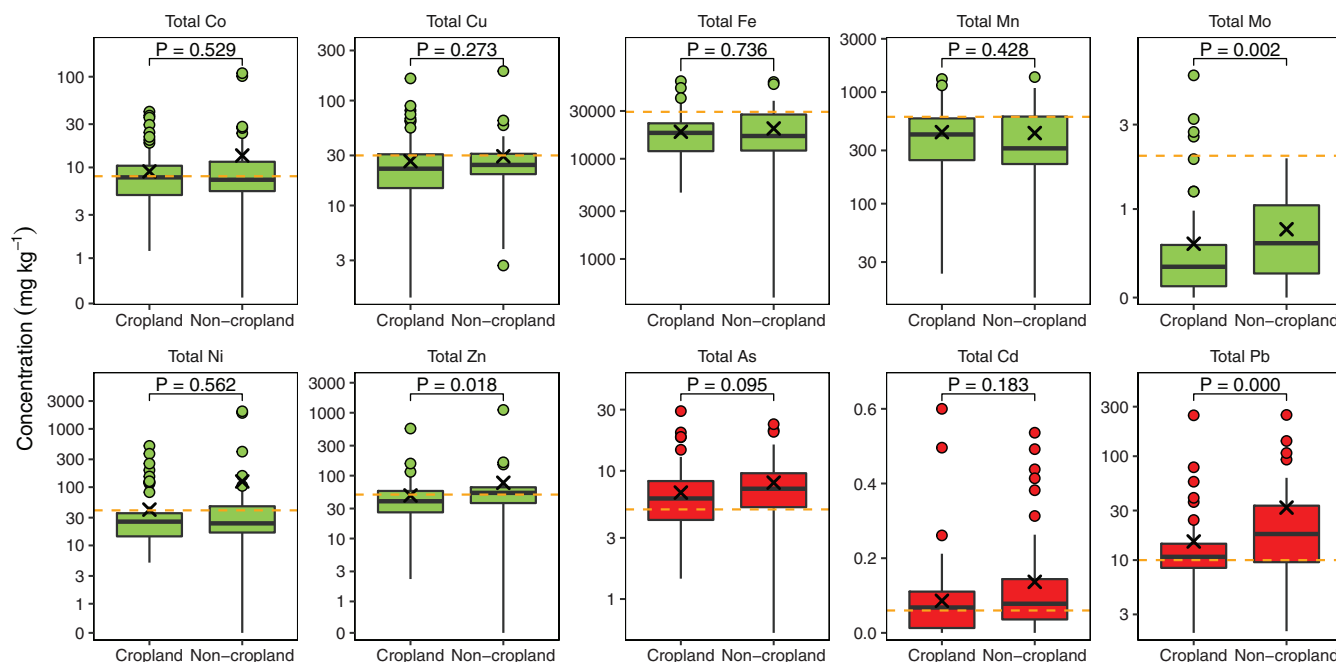
concentration of contaminants, especially that of As and Cd, was slightly above typical levels (Figure 1). This last result may be a reflection of a potential diffuse contamination due to the historical anthropogenic impact on European soils (Nriagu, 1996), and particularly to the application of mineral fertilisers, which are a major source of these toxic elements in soil (Atafar et al., 2010; Chen et al., 2008). Nonetheless, the total concentrations of the elements examined here tended to be smaller for croplands than for non-croplands, especially for Mo ( $p < 0.01$ ), Zn ( $p < 0.05$ ) and Pb ( $p < 0.001$ ) and we observed no evidence that agricultural activity posed an obvious input of contaminants to soils.

Micronutrient and contaminant uptake by crops is usually related to the so-called available fraction, which is usually determined by DTPA-like extractions, rather to the soil total content (McLaughlin et al., 2000). Broadly established thresholds of micronutrient availability, below which crop production may be negatively affected, are  $4.5 \text{ mg kg}^{-1}$  for Fe,  $1 \text{ mg kg}^{-1}$  for Mn,  $0.5 \text{ mg kg}^{-1}$  for Zn and  $0.2 \text{ mg kg}^{-1}$  for Cu (Li et al., 2007; Lindsay & Norvell, 1978). Considering these values, we found 60% of the dryland European soils in the suboptimal range for Fe, 28% for Zn and <1% for Mn, while all soils showed available Cu concentrations above the deficiency threshold (Figure 2). It is known that many European agricultural soils need to be supplied with micronutrients (Agriculture and Rural Development, 2019; Ballabio et al., 2018) but, so far, there has been no systematic and large-scale quantification of these deficiencies. When comparing soil management, we found that soil available micronutrients Zn and Mo were significantly lower in cropland than in non-cropland soils ( $p < 0.001$  and  $< 0.01$  respectively), as well as contaminant Pb ( $p < 0.01$ ).

#### 3.2 | Drivers of bioavailability of soil micronutrients and contaminants

We studied the contribution of soil properties (clay, pH, organic C, and total element) and aridity to the availability of micronutrients and contaminants using confirmatory path analysis (CPA, Figure S3). For this purpose, we examined the relative importance of each soil and environmental variable, first on the standardised averages of the available concentrations of all micronutrients and all contaminants, and then on the available concentrations of each element separately.

Our data fitted very well the selected CPA models (Fisher C  $p$  value > 0.73) and explained 51% of the variance of the standardised mean micronutrient availability and 31% of the standardised mean contaminant availability (Figure 3). We found positive effects of total



**FIGURE 1** Boxplots of total concentrations of soil micronutrients (green) and contaminants (red) in croplands ( $n = 103$ ) and non-croplands ( $n = 45$ ) of European drylands. Linear scale from 0 to 1  $\text{mg kg}^{-1}$  and logarithmic scale from 1  $\text{mg kg}^{-1}$ . Crosses indicate means. Dashed horizontal lines indicate estimated global averages (Lindsay, 1979; Zuo & Zhang, 2011).  $p$ -values of unpaired two-sample Wilcoxon rank-sum tests comparing element concentrations of croplands versus non-croplands are shown in the graphs [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

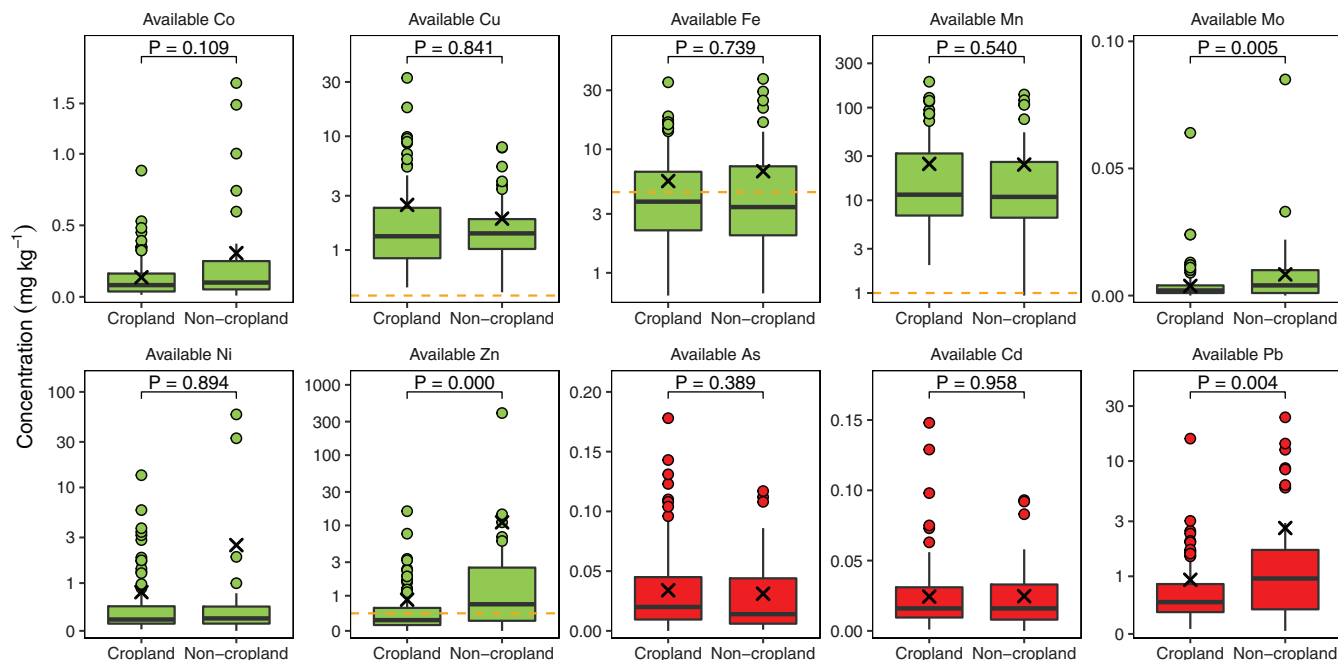
micronutrient and organic C concentrations and strong negative effects of pH on the availability of micronutrients. In contrast, the availability of contaminants was only affected by their total concentration.

For each specific micronutrient, our models explained 88% of the variance of available Ni, 80% of the variance of available Zn, 64% of the variance of available Mn, 58% of the variance of available Fe, 58% of the variance of available Co, 30% of the variance of available Mo, and 27% of the variance of available Cu (Figure S4). Available Fe was positively influenced by total Fe and organic C and negatively influenced by pH (very strong negative influence) and aridity (Figure 4). Available Zn was positively and strongly influenced by total Zn, and negatively by clay content. Similarly, Ni availability was strongly and positively influenced by total Ni and slightly and negatively by pH. Available Mn was positively affected by total Mn, but negatively by pH, while organic C had a positive total effect (sum of direct and indirect effects). Available Co was positively influenced by total Co and organic C and negatively by pH, but total Cu was positively affected by clay content and negatively by pH. Finally, available Mo was positively related to total Mo and negatively influenced by pH and clay content.

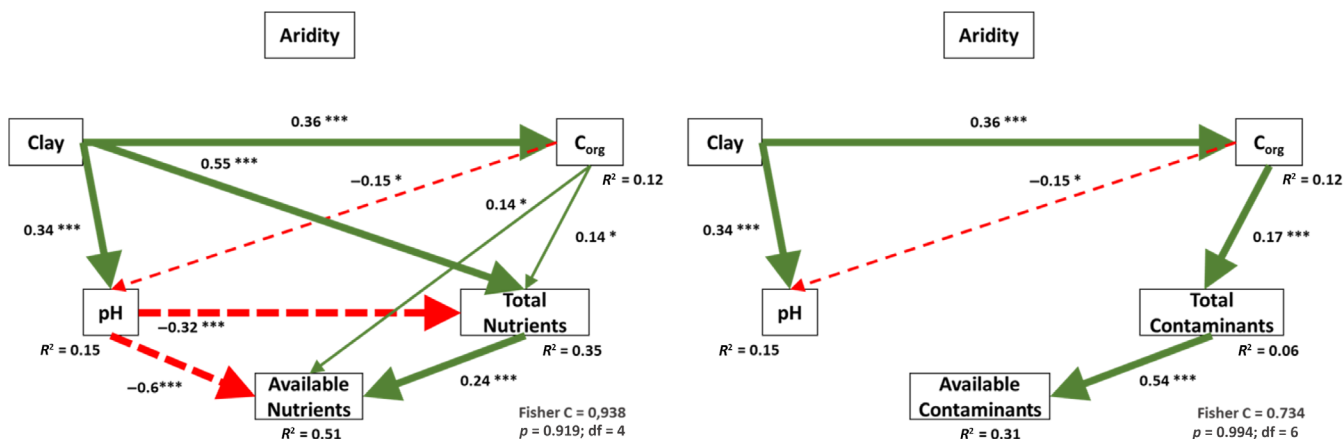
In summary, the availability of all micronutrients was mostly affected by their total concentrations (Figure 4). Furthermore, most available micronutrients were negatively affected by pH, which may be due to the formation of insoluble species and immobilising chemical reactions promoted by pH rise (McBride, 1989; Sauvé et al., 2000). The only exceptions were Cu and Zn availability. Organic C often positively affected both available and total metallic micronutrient contents through direct and indirect effects, probably because many of these elements are, to some extent, associated with soil organic matter (Carter & Stewart, 1995; Loveland & Webb, 2003; Wichard et al., 2009). Clay content positively influenced some of the metal availabilities in soils, namely Co, Cu, Fe and Mn, but not Mo, Ni or Zn.

Regarding the contaminants (Figure S5), the models explained 41% of the variance of available Cd, 38% of available Pb and 27% of available As. Available Cd was negatively affected by pH and aridity and positively by total Cd and clay content. Available Pb was influenced directly by its total concentration. Finally, available As was positively influenced by clay content and negatively by pH. Clay content had a positive total association to As and Pb availability and organic matter seems to enhance available contaminants in soils (Figure 4), probably





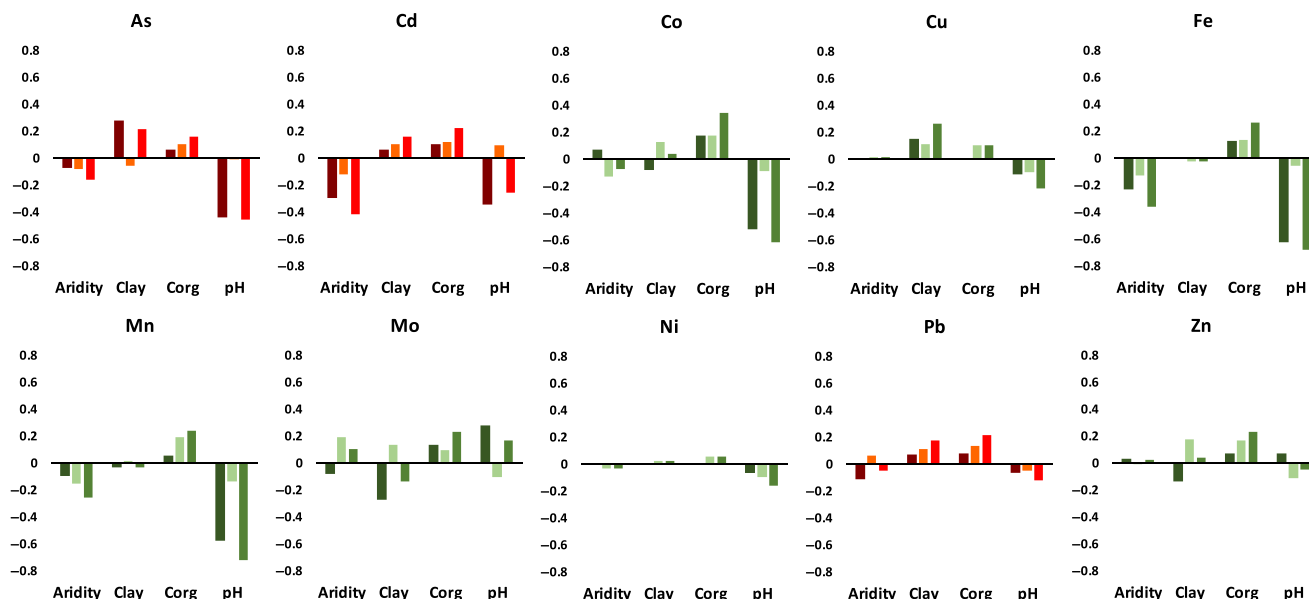
**FIGURE 2** Boxplots of available (DTPA-extractable) concentrations of soil micronutrients (green) and contaminants (red) in croplands ( $n = 103$ ) and non-croplands ( $n = 45$ ) of European drylands. Linear scale from 0 to  $1 \text{ mg kg}^{-1}$  and logarithmic scale from  $1 \text{ mg kg}^{-1}$ . Crosses indicate means. Dashed horizontal lines show DTPA-extractable Cu, Fe, Mn and Zn deficiency limits for crops (Li et al., 2007; Lindsay & Norvell, 1978).  $p$ -values of unpaired two-sample Wilcoxon rank-sum tests comparing element concentrations of croplands versus non-croplands are shown in the graphs [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]



**FIGURE 3** Effects of aridity, clay percentage, pH, organic carbon, and total content on available micronutrients and contaminants. Left, micronutrients. Right, contaminants. Numbers adjacent to arrows are standardised path coefficients (analogous to relative regression weights) and indicative of the effect of the relationship. Continuous green arrows show positive relationships and dashed red arrows show negative relationships, with arrow thicknesses proportional to the strength of the relationship. The proportion of variance explained ( $R^2$ ) is shown beneath each response variable in the model. Goodness-of-fit statistics are shown in the lower right corner in grey. The a priori model was refined by removing paths with non-significant relationships (see the a priori model in Figure S3). \* $p < 0.05$ ; \*\* $p < 0.01$ ; \*\*\* $p < 0.001$  [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

because these contaminants may be associated with soil organic matter (Martín et al., 2006; Micó et al., 2006; Park et al., 2011; Qishlaqi & Moore, 2007). Alkaline soil pH, on the other hand, was associated with lower

concentrations of Cd and As. Cd availability is known to decrease upon the increase of soil pH (Sauvé et al., 2000). However, rising pH is usually associated with higher As availability (Moreno-Jiménez et al., 2012). Exploring this



**FIGURE 4** Standardised direct (dark), indirect (light) and total sum of effects (intermediate) of aridity, clay content, organic carbon (Corg) and pH on available metal concentrations, as derived from the confirmatory path analysis. Micronutrients in green tones, contaminants in red tones [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

into detail, available As exhibited a quadratic relationship with pH, with a maximum at pH  $\sim 7.4$  (Figure S6). The decrease of available As at higher pH may be associated to concurring factors not taken into account in our models, such as the parent material; also, unlike for the rest of the elements examined, DTPA extraction may reflect poorly the availability of As in soils.

#### 4 | IMPLICATIONS FOR MANAGING EUROPEAN SOILS

The actions taken to ensure food security and quality and those to avoid soil contamination and protect human health often do not coincide (Kopittke et al., 2019). This is particularly true for agricultural soils and in an overall scenario of global warming. As the role of metals in soil is diverse, as nutrients or contaminants, a trade-off between positive (provided by nutrients) and negative (provided by contaminants) effects, and thus between food production and soil conservation, needs to be found. Our work moves in this direction by providing observational evidence of the influence of geochemical and aridity properties on metal availability in European dryland soils and, thus, proposing measures to be taken to enhance plant nutrient uptake and reduce contamination risk. This is of particular interest considering that European dryland soils are mostly devoted to food and plant production (agriculture, pastures or forests) (Gallego & Bamps, 2008).

In this respect, we identify Fe as the main deficient element in soil, and Zn in some cases. From a geochemical perspective, this agrees with the soil properties typically found in dryland soils of southern Europe, namely high pH and low organic matter contents. As a result, micronutrient fertilisation in these areas is very common to maintain good levels of production (Eurostat, 2012). Nonetheless, this practice contrasts with the actions recently proposed as part of the European Green Deal by the European Commission (From Farm to Fork Strategy), which aim to reduce fertiliser use by at least 20% by 2030 to promote a more sustainable EU food system (COM/2020/381, Farm to Fork Strategy for a fair, healthy and environmental-friendly food system, 2020). In light of our findings, here we propose some specific recommendations that may contribute to achieve such an ambitious goal at EU level. Interestingly, we found that the availability of micronutrients such as Zn, Mo and, to a lesser extent, Co were lower in croplands, suggesting the stripping of micronutrients from soil during continuous cropping in agricultural soils. Future policies should also consider this trend because it may threaten the nutritional stability of the agro-system.

First, our study confirms a limited number of potential actions to ensure a satisfactory micronutrient supply for plant growth and simultaneously avoid contamination. Careful management practices should be put in place by farmers in order to precisely act on those soil properties that can ensure positive (i.e., micronutrient availability enhancement) and reduce negative (i.e., contamination risk

increase) effects. Indeed, farmers usually tend to add acidic amendments to calcareous soils to lower pH and increase micronutrient availability (Starast et al., 2007). In agreement with our findings, this measure may be effective. Nonetheless, farmers should be careful with concomitant increases in the availability of Cd, which acts as contaminant and could enter into the food chain (Cui et al., 2004).

In addition, modern agricultural practices rely on conserving and building up soil organic C (Fageria, 2012; Wolf & Snyder, 2003) that, according to our results, may improve soil micronutrient availability. The organic amendments used for this purpose should be of good quality, with low content of metallic contaminants (Alvarenga et al., 2015; Smith, 2009), because potential increases in total As, Cd and Pb may enhance the respective available pools with the subsequent risk for crop and water contaminations. We show that clay content is also important for soil metal retention and, thus, for promoting their slow release into the available pools (Fageria et al., 2002). However, practices for managing clays are more complicated than those acting on pH or organic matter, thus difficult to be applied at large scale.

Overall, our findings support the need for a strategy based on ad-hoc agricultural actions that take into account the peculiarities of each area, including different factors (e.g., climate and biodiversity) and putting soil at the core of the entire process. In the specific case of dryland soils, management practices that promote the reduction of fertiliser use and minimise the contamination risk by acting on soil pH and organic matter content should be considered when setting up farmer subsidy schemes as part of the EU agricultural policy. The same measures might not be effective in other regions with soils with other properties. Therefore, studies similar to ours on areas with different properties would generate and make available a reliable large-scale assessment to decision-makers.

In addition to this, any agricultural planning and actions must consider the overall environmental conditions. In particular, as shown here, climate may also have an impact on soil biogeochemistry. Indeed, the arid conditions in drylands, currently located mainly in southern Europe but likely expanding in the future, will tend to increase in intensity over the coming decades (higher temperatures and less water availability) (Cramer et al., 2018). In this context, we found that Fe is negatively affected by aridity, in agreement with a previous global observation in non-agricultural soils (Moreno-Jiménez et al., 2019). Mechanisms behind this sensitivity of available Fe to aridity remain unsolved, although they may be related to the several (hydr)oxides that this metal can form in soil (Churchman & Lowe, 2012; Kämpf & Schwertmann, 1983). Most concerning is our finding that in the European drylands this sensitivity is accompanied

by an Fe availability often below the optimal levels for crops (in ~60% of the analysed soils). This may represent both agroenvironmental and human health issue.

First, Fe fertilisation, mainly relying on a predominant use of synthetic fertilisers from non-renewable resources (Shuman, 1998), is widespread in agriculture (Brown, 2008). Nonetheless, its efficacy depends on environmental factors. For instance, the availability of micronutrients may become unbalanced as each shows different sensitivity to aridity (e.g., available Zn is less affected than Fe). The potential uncoupling of micronutrients has received much less attention compared to that of macronutrients (C, N, P and K) (Sardans & Peñuelas, 2015; Zheng et al., 2021), yet these imbalances may have serious agroecological consequences. Our study clearly shows, for the first time, the risk of micronutrient uncoupling occurring in dryland soils of Europe.

Finally, our results show that the foreseen rise in aridity can result in possible serious food security and human health issues. Indeed, an increment in aridity may intensify Fe starvation in crops and lead to serious reductions in the average human intake of this element, a nutrient that is already deficient in our diet (Thompson & Amoroso, 2010). Such tendencies have been already reported for Fe and Zn in wheat between 1970 and 2000 (Zhao et al., 2009). In light of our results, the registered increase of aridity in this period may have been a cause of the detected micronutrient reduction in wheat (Zarch et al., 2015). More than a future risk scenario, crop Fe deficiency seems to be an issue already occurring in agricultural fields in Europe.

So far, alternatives to N and P synthetic fertilisers for a more sustainable agricultural production have received a great deal of attention, mainly pushed by their worrying scarcity and the environmental issues associated with the exploitation of N and P sources (Dawson & Hilton, 2011; Schoumans et al., 2015). In contrast, the search for sustainable alternative sources of micronutrients has been neglected (Bindraban et al., 2015). Our data suggest that current agricultural schemes will not be enough to overcome soil Fe limitations; thus, soon we may urge alternatives to efficiently and sustainably face an Fe deficiency in crop production.

In conclusion, for any agro-ecosystem, when setting up actions for ensuring agricultural productivity and simultaneously reducing the use of fertilisers (as proposed by the EU Farm to Fork Strategy), specific soil (e.g., pH and organic C content) and climate (e.g., aridity risk) parameters have to be put at the core of any decision process to supply scarce micronutrients to crops. A trade-off approach, based on ad hoc measures, has shown to be an adequate way to ensure a sustainable management of soil (preserve the availability of



micronutrients in soil), reduce environmental impacts (mitigate contaminants) and guarantee a production of good quality food for human health care (maximising nutritional status while preventing food contamination).

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## CONFLICT OF INTEREST

The authors declare no competing interests.

## DATA AVAILABILITY STATEMENT

The data used in analyses are available from figshare (<https://figshare.com/s/3f69be6fc92c1edd1e3d>).

## AUTHOR CONTRIBUTIONS

**Eduardo Moreno-Jimenez:** Conceptualization (lead); data curation (equal); formal analysis (equal); funding acquisition (lead); investigation (lead); methodology (supporting); project administration (lead); resources (equal); supervision (lead); visualization (equal); writing – original draft (lead); writing – review and editing (equal). **Alberto Orgiazzi:** Conceptualization (supporting); data curation (equal); formal analysis (supporting); investigation (supporting); methodology (equal); resources (equal); supervision (supporting); writing – original draft (supporting); writing – review and editing (equal). **Arwyn Jones:** Data curation (equal); methodology (supporting); resources (equal); writing – review and editing (supporting). **Hugo Saiz:** Conceptualization (supporting); data curation (supporting); formal analysis (equal); software (lead); writing – review and editing (equal). **Sara Aceña Heras:** Data curation (supporting); formal analysis (supporting); investigation (supporting); methodology (lead); writing – review and editing (supporting). **César Plaza de Carlos:** Conceptualization (supporting); data curation (supporting); formal analysis (equal); investigation (equal); supervision (supporting); visualization (equal); writing – original draft (equal); writing – review and editing (equal).

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